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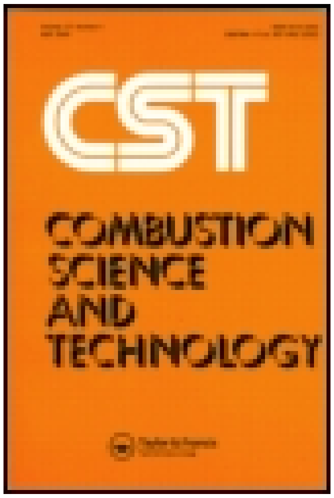

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A Regime Diagram for Autoignition of Homogeneous Reactant Mixtures with Turbulent Velocity and Temperature Fluctuations

Short title: Regime Diagram for Autoignition

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ABSTRACT (150 words)

A theoretical scaling analysis is conducted to propose a diagram to predict weak and strong ignition regimes for a compositionally homogeneous reactant mixture with turbulent velocity and temperature fluctuations. The diagram provides guidance on expected ignition behavior based on the thermo-chemical properties of the mixture and the flow/scalar field conditions. The analysis is an extension of the original Zeldovich's analysis by combining the turbulent flow and scalar characteristics in terms of the characteristic Damköhler and Reynolds numbers of the system, thereby providing unified and comprehensive understanding of the physical and chemical mechanisms controlling ignition characteristics. Estimated parameters for existing experimental measurements in a rapid compression facility show that the regime diagram predicts the observed ignition characteristics with good fidelity.

Keywords: ignition regimes, scaling analysis, weak/strong ignition, temperature fluctuations

1. INTRODUCTION

Towards clean and efficient energy utilization, new strategies in combustion devices for both automotive and stationary applications operate using lean, nearly homogeneous reactant mixtures at boosted pressure and preheated conditions. These include aircraft engines operating at higher inlet temperatures (Lieuwen et al., 2013), low temperature combustion engines (Lavoie et al., 2010), and stationary gas turbines using lean premixed combustion (US DOE, 2009), among many examples. Under these conditions, autoignition often becomes a dominant process for burning. As such, accurate prediction of autoignition characteristics – the ignition delay times as well as the entire evolution of the fuel consumption behavior – is of paramount importance in successful implementation of the combustion systems.

Historically, the subject of autoignition and slow versus rapid combustion has been extensively studied in the context of detonation and explosion research. Earlier experimental studies of shock-induced ignition reported distinct ignition behavior, referred to as the “weak” and “strong” ignition regimes, for which the importance of the ignition delay time sensitivity, $d\tau_{ig}/dT$, was recognized (Meyer and Oppenheim, 1971; Oran et al., 1982; Oran and Boris, 1982). It was found that the $d\tau_{ig}/dT$ iso-lines serve as a rational criterion to distinguish the different ignition regimes. A further theoretical study by Zeldovich (1980) proposed criteria for ignition regimes, classified as detonation, spontaneous propagation, and normal flame. The issue of premature ignition by local hot spots within a shock tube has also been revisited in recent studies (Uygun et al., 2014; Javed et al., 2015).

Recognizing the general interest in fundamental characterization of autoignition phenomena, the present study was further motivated by the recent research activities on combustion of coal-derived syngas in gas turbines as a new strategy for clean utilization of coal (US DOE, 2009). Due to the high flame temperatures and NO_x emissions associated with high hydrogen content fuels like syngas, a common combustion strategy is to operate in the lean premixed mode (Richards et al., 2001). In such conditions, combustion stability depends more highly on autoignition characteristics (Lieuwen et al., 2008). As for the autoignition characteristics of syngas mixtures at typical gas turbine operating conditions (20 bar and above), a compilation of recent experimental and computational studies was reported in Petersen et al. (2007), showing a wide range of discrepancies between measurements and predictions based on homogeneous adiabatic calculations with detailed chemistry, especially at low temperature (< 1000 K) conditions.

Subsequently, a number of studies followed in order to identify the main cause of such discrepancies. Experimental studies using rapid compression facilities (RCF) reported a possibility that the discrepancies may be attributed to the non-uniform temperature and mixture fields arising from wall heat loss and flow vortex generation (Mittal et al., 2006; Walton et al., 2007). A recent study by Mansfield and Wooldridge (2014) conducted imaging experiments of syngas autoignition within an RCF, and reported an early phase front propagation, called the "weak" ignition regime, at low temperature conditions. They also confirmed that the ignition delay in such conditions is significantly shorter, by several factors, than the corresponding homogeneous ignition delay prediction. A pressure-temperature diagram was provided to distinguish between the weak and strong ignition regimes. Moreover, several criteria to identify the transition between weak and strong ignition regimes were evaluated, including the criterion by Zeldovich (1980). The modified formula proposed by Sankaran et al. (2005), based (in part) on the ignition delay time sensitivity, $d\tau_{ig} / dT$, was found to reproduce the experimentally observed trends very well. To corroborate experimental findings, Ihme and co-workers (Ihme, 2012; Wu and Ihme, 2014) used simple one- and two-dimensional models to demonstrate that the presence of turbulent fluctuations can lead to significant ignition advancement. These recent series of findings have led to a consensus in the community that scalar non-uniformities are the likeliest causes of the discrepancies between zero-dimensional modeling and the experimental autoignition delay time data (US DOE UTSR, 2014).

Considering the established significance of thermal and compositional non-uniformities on autoignition characteristics, the main objective of the present work is to extend the understanding to develop a rational criterion to predict the conditions associated with the different autoignition regimes for general turbulent mixing conditions. To this end, the diagrams shown in the experimental studies (Mansfield and Wooldridge, 2014; Meyer and Oppenheim, 1971), which focused on a regime criterion solely based on chemical characteristics of the ignition sensitivity, are not sufficient; such a criterion lacks other potentially important information about the level of scalar fluctuations which trigger the front initiation and propagation.

To elaborate further, the Zeldovich criterion (Zeldovich, 1980), which was further refined and demonstrated by Sankaran et al. (2005), defines the non-dimensional number, Sa, as

$$\mathrm{Sa} = \beta \frac{S_L}{S_{sp}} = \beta S_L \left| \frac{d\tau_{ig}}{dT} \nabla T \right|, \quad \beta = 0.5 \tag{1}$$

where S_L is the laminar flame speed, $S_{sp} = \left| \frac{d\tau_{ig}}{dT} \nabla T \right|^{-1}$ is the spontaneous ignition front propagation speed, and τ_{ig} is the ignition delay time for the homogeneous mixture at the average or bulk temperature. The factor $\beta < 1$ reflects the fact that sufficiently rapid spontaneous front propagation is needed in order to ensure strong ignition. Hereinafter, Equation (1) will be referred to as the *Zeldovich-Sankaran* criterion. The ignition regime criterion predicts weak ignition if Sa > 1, as the deflagration front dominates the ignition behavior, and strong ignition if Sa < 1, in which case the spontaneous ignition process dominates. Therefore, it is evident that the ignition regimes are determined by the ignition delay sensitivity $(d\tau_{ig} / dT)$ and the temperature distribution (∇T), which is determined by the scalar field distribution. In RCF and shock tube autoignition studies of syngas, thermal gradients on the order of 5 K/mm are expected (Mansfield and Wooldridge, 2014); however, systematic studies of the effects of flow and scalar field fluctuations are needed to expand the predictive regime diagram to realistic combustion devices where much larger temperature gradients can be expected.

This study presents a theoretical scaling analysis to extend the regime criterion in terms of non-dimensional parameters that are commonly used in characterizing turbulent combustion systems. In the following, the relevant physical quantities are identified and simplifying assumptions are introduced. Subsequent derivations of relevant scaling relations then lead to the ignition regime criterion with turbulent combustion parameters. The predicted regime diagram is then validated by the evaluation of the conditions encountered in experimental measurements.

2. PROBLEM DEFINITION AND ASSUMPTIONS

Figure 1 shows a schematic of the problem under consideration, and important characteristic quantities. The length scales include the chamber length, *L*, the integral eddy scale, ℓ, the Taylor microscale, λ, and the deflagration flame thickness, δ_f. In general, ℓ is considered a fraction of *L*, and λ/ℓ scales with the turbulent Reynolds number as will be discussed later. The laminar flame speed, S_L, and the root-mean-square (RMS) turbulent velocity fluctuation at the integral scale, u', are important velocity scales that will be compared to the other relevant velocities to be determined later. For the scaling analysis, the following simplifications and assumptions are made:

1. Weak ignition is primarily caused by front propagation originating from small scale local temperature fluctuations, with a length scale typically of an order of mm or less, such as

local hot spots, and the effects of large scale bulk temperature gradients, such as gradients caused by wall heat losses, are not considered. This is based on the experimental observations that early stage ignition kernels are often generated in the interior of the combustor, not necessarily near the wall region.

2. The mixture composition is homogeneous, and only the temperature fluctuations are considered. The scales of initial temperature and velocity fluctuations are comparable and correlated.

3. The Prandtl number of the mixture is unity, so that combined with assumption 2, the dissipation of temperature fluctuations is mainly driven by turbulent flows. This implies that the time and length scales for turbulent velocity and scalar fields are the same (i.e. the Batchelor scale is identical to the Kolmogorov scale).

To characterize the turbulent velocity and scalar fields, key non-dimensional parameters are introduced. Following the framework of Liñán and Williams (1993), a rational way to characterize turbulent combustion systems is to use the turbulent Reynolds number, which represents the intensity of turbulence, and the characteristic turbulent Damköhler number, which represents the intensity of chemical reaction. For the integral scale eddy whose velocity, length, and time scales are characterized by u', ℓ, $\tau_\ell = \ell / u'$, respectively, the turbulent Reynolds number is defined as:

$$\mathrm{Re}_\ell = \frac{u'\ell}{\nu} \tag{2}$$

where ν is the kinematic viscosity of the bulk mixture gas. As for the measure of the chemical intensity, two ignition Damköhler numbers are defined as:

$$\mathrm{Da}_{\ell} = \frac{\tau_{\ell}}{\tau_{ig}} \tag{3}$$

which is referred to as the *integral* Damköhler number, and

$$\mathrm{Da}_{\lambda} = \frac{\tau_{\lambda_T}}{\tau_{ig}} \tag{4}$$

is referred to as the *mixing* Damköhler number, where τ_{ig} is the ignition delay time for the homogeneous reactant mixture at the bulk temperature, τ_{λ_T} is the mixing time scale associated with the Taylor microscale for the temperature field, λ_T, which are determined in terms of the RMS temperature fluctuation, T', and the mean temperature dissipation rate, $2\alpha\widetilde{|\nabla T|^2}$, written as

$$\tau_{\lambda_T} = \frac{T'^2}{\alpha\widetilde{|\nabla T|^2}},\ \lambda_T^2 = \frac{T'^2}{\widetilde{|\nabla T|^2}} \tag{5}$$

in analogy with those of the Taylor microscales for velocities

$$\tau_{\lambda} = \frac{u'^2}{\nu\widetilde{|\partial u_j / \partial x_j|^2}},\ \lambda^2 = \frac{u'^2}{\widetilde{|\partial u_j / \partial x_j|^2}} \tag{6}$$

Based on assumption 3, it follows that the mixing time and length scales for temperature are interchangeable with those for turbulent velocities, such that

$$\tau_{\lambda_T} = \tau_{\lambda},\ \lambda_T = \lambda\,. \tag{7}$$

3. SCALING ANALYSIS

The main objective of the scaling analysis is to derive an expression for the Zeldovich-Sankaran criterion in terms of the characteristic Reynolds and Damköhler numbers. Recalling from the theory of homogeneous turbulence (Tennekes and Lumley, 1972), the scaling relation yields:

$$\frac{\lambda}{\ell} = \mathrm{Re}_{\ell}^{-1/2}, \ \frac{\tau_{\lambda}}{\tau_{\ell}} = \frac{\lambda / u'_{\lambda}}{\ell / u'} = \frac{\lambda}{\ell}\frac{u'_{\lambda}}{u'} = \left(\frac{\lambda}{\ell}\right)^{2/3} = \mathrm{Re}_{\ell}^{-1/3}. \qquad (8)$$

It follows that

$$\mathrm{Da}_{\lambda} = \frac{\tau_{\lambda_T}}{\tau_{\mathrm{ig}}} = \frac{\tau_{\lambda}}{\tau_{\mathrm{ig}}} = \frac{\tau_{\ell}}{\tau_{\mathrm{ig}}}\frac{\tau_{\lambda}}{\tau_{\ell}} = \mathrm{Da}_{\ell}\,\mathrm{Re}_{\ell}^{-1/3}, \qquad (9)$$

The significance of Da_{λ} is that it is the ratio of the characteristic temperature dissipation time to the characteristic ignition delay time at the bulk mean temperature. Therefore, if $\mathrm{Da}_{\lambda} < 1$, the temperature fluctuations are dissipated before ignition occurs, thus it is unlikely to exhibit the weak ignition behavior triggered by reaction front propagation.

The next step is to extend the Zeldovich-Sankaran criterion, Equation (1), to turbulent conditions. To this end, it is assumed that the occurrence of the hot-spot-induced pre-ignition is proportional to the statistical mean temperature gradient, such that

$$\mathrm{Sa} \approx \beta S_L \left|\frac{d\tau_{ig}}{dT}\right| \widetilde{|\nabla T|} \qquad (10)$$

where it is estimated that

$$\left|\widetilde{\nabla T}\right| \approx \frac{T'}{\lambda_T} \approx \frac{T'}{\lambda} = \frac{T'}{\ell\,\mathrm{Re}_\ell^{-1/2}} \tag{11}$$

Therefore, Equation (10) is written as

$$\mathrm{Sa} \approx \beta S_L \left|\frac{d\tau_{ig}}{dT}\right| \frac{T'}{\ell}\mathrm{Re}_\ell^{1/2} = \beta\left(\frac{S_L}{\delta_f}\right)\left(\frac{\delta_f}{\ell}\right)T'\left|\frac{d\tau_{ig}}{dT}\right|\mathrm{Re}_\ell^{1/2} \tag{12}$$

which includes the length scale ratio, δ_f/ℓ where δ_f is the characteristic flame thickness. Following Liñán and Williams (1993),

$$\frac{\delta_f}{\ell} = \mathrm{Re}_\ell^{-1/2}\,\mathrm{Da}_{\ell,f}^{-1/2} = \mathrm{Re}_\ell^{-1/2}\left(\frac{\tau_\ell}{\tau_f}\right)^{-1/2} = \mathrm{Re}_\ell^{-1/2}\left(\frac{\tau_\ell}{\tau_{ig}}\right)^{-1/2}\left(\frac{\tau_{ig}}{\tau_f}\right)^{-1/2} = \mathrm{Re}_\ell^{-1/2}\,\mathrm{Da}_\ell^{-1/2}\left(\frac{\tau_{ig}}{\tau_f}\right)^{-1/2}, \tag{13}$$

where it is noted that the integral Damköhler number in Liñán and Williams (1993) was defined differently from Eq. (3) above, and was based on the flame time scale, $\tau_f = \delta_f / S_L$. Therefore, the factor τ_{ig}/τ_f must be included. Combining Equations (12) and (13), the turbulent ignition regime criterion can be written as:

$$\mathrm{Sa} = K\mathrm{Da}_\ell^{-1/2},\ K = \beta\left(\frac{T'}{\left(\tau_f\tau_{ig}\right)^{1/2}}\right)\left|\frac{d\tau_{ig}}{dT}\right| \tag{14}$$

where K is referred to as the normalized thermal ignition sensitivity. In comparison with the laminar version in Eq. (1), $S_L\left|\nabla T\right|$ has now been expressed as $\left(T'/\tau^*\right)\mathrm{Da}_\ell^{-1/2}$ through the dimensional scaling, with a *reduced* time scale, $\tau^* = \left(\tau_f\tau_{ig}\right)^{1/2}$. The final ignition regime criterion becomes:

$$\begin{cases}\mathrm{Da}_\ell < K^2: \text{ weak ignition}\\ \mathrm{Da}_\ell > K^2: \text{ strong ignition}\end{cases} \tag{15}$$

As discussed with Equation (9), an additional condition of $\mathrm{Da}_\lambda = \mathrm{Da}_\ell \mathrm{Re}_\ell^{-1/3} > 1$ needs to be satisfied to ensure weak ignition, since otherwise the temperature fluctuations are likely to dissipate away before the front forms. Finally, $\mathrm{Da}_\ell < 1$ would ensure an even stronger mixing scenario, since eddies at all scales would have time scales shorter than the ignition delay time, such that all temperature fluctuations would be dissipated and only strong ignition would be expected.

4. THE REGIME DIAGRAM AND DISCUSSION

Compiling the above scaling analysis leads to the regime diagram as shown in Figure 2. The autoignition processes in nearly homogeneous mixtures with turbulent fluctuations are characterized in the Da-Re space, to represent the relative chemical and turbulence intensities determined by the chemistry, thermodynamics and turbulent transport in the gas mixture. It is shown that the primary factor to determine the ignition regime is Da_ℓ, while Re_ℓ modifies the conditions further.

First, for a given Re_ℓ, the Zeldovich-Sankaran criterion indicates that the weak/mixed ignition regime is possible for $1 < \mathrm{Da}_\ell < K^2$. If $\mathrm{Da}_\ell > K^2$, then the reactant mixture is either too reactive (small τ_{ig}) or the mixture ignition characteristics are not sensitive to the temperature fluctuations (small $d\tau_{ig} / dT$), such that the entire mixture ignites almost at the same time despite some level of temperature fluctuations. This is referred to as the *reaction-dominant* strong ignition regime. On the other hand, if $\mathrm{Da}_\ell < 1$, then the turbulent mixing is rapid (small τ_ℓ) such that the temperature fluctuations are dissipated before the local ignition takes place. In contrast to the $\mathrm{Da}_\ell > K^2$ case, this is referred to as the *mixing-dominant* strong ignition regime. Note that the K parameter includes the ignition delay sensitivity, which is more than just a time scale characterization, and depends strongly on the ignition chemistry of the specific fuel.

Between the limits $1 < \mathrm{Da}_\ell < K^2$, weak ignition is possible; however, the mixing Damköhler number, Da_λ, provides an additional criterion for this region of the regime diagram. Considering that the dynamics of turbulent mixing and dissipation is commonly characterized by the Taylor scale, λ, a proper criterion to determine the mixing-dominant strong ignition would be the ratio of the Taylor mixing time, τ_λ, to the ignition time, τ_{ig}. Therefore, the $\mathrm{Da}_\lambda = 1$ condition serves as a more refined criterion within the limits of $1 < \mathrm{Da}_\ell < K^2$ to further identify the boundary between the weak and strong ignition regimes. Considering Equation (9), this line appears on the regime diagram with a slope of 1/3, indicating that the occurrence of weak ignition phenomena will become less likely as the turbulent Reynolds number of the mixture increases. Still, the conditions between $\mathrm{Da}_\lambda < 1$ and $\mathrm{Da}_\ell > 1$ are a "grey" zone, in that some mixed mode ignition in which a mild level of front propagation followed by a strong ignition may occur. This is denoted as the *mixing-dominant mixed/strong* ignition regime.

The regime diagram serves as a qualitative guidance to the expected ignition behavior. The appropriate autoignition regime can be identified given the knowledge of the thermo-chemical properties of the mixture (e.g. pressure, temperature, reaction chemistry, etc.), and the characteristic turbulent flow parameters (e.g. Reynolds number, turbulence/scalar fluctuation intensity, etc.). If the initial condition of the mixture falls into the weak ignition or mixed/strong ignition regimes, then large discrepancies in the ignition delay prediction against the measured data can be expected and must be treated carefully.

To validate that the proposed regime diagram predicts the ignition characteristics in actual systems, the experimental data for syngas autoignition by Mansfield and Wooldridge (2014) are processed and plotted on the regime diagram, as shown in Figure 3. The homogeneous ignition delay times are computed using CHEMKIN (Kee et al., 1989) calculations with the detailed reaction mechanism by Li et al. (2007). The experimental conditions, the types of the observed ignition behavior and the corresponding regime diagram parameters are provided in Table 1. The turbulence parameter estimation was difficult, as no detailed measurements of flow field fluctuations were available, as is often the case with many ignition experiments. Therefore, estimations were made based on the reported mean velocity and presumed level of turbulence intensity at 1%, turbulence integral length scale, $\ell \sim 0.4L$, (where L is the dimension of the combustion chamber) and a temperature fluctuation, $T' \sim 10$ K. Since the K^2 line also depends on the physico-chemical parameters of the mixture, each experimental data point yields a separate horizontal line on the regime diagram.

Figure 3(a) and (b) show a compilation of selected data points corresponding to the strong and weak ignition behavior, respectively. Although not all available data points are shown in order to avoid clutters, it was confirmed that most of the data points represented correct ignition behavior predicted by the regime diagram. While there are some uncertainties in the parameter estimations (especially those related to turbulent fluctuations), the experimental data points mapped on the regime diagram are found to be in very good agreement with the proposed theory. More experimental data with detailed flow field measurements will be needed to validate the theoretical prediction for a wide range of device and mixture conditions.

5. CONCLUDING REMARKS

A theoretical scaling analysis was conducted to develop a regime diagram to predict weak and strong ignition regimes for a compositionally homogeneous reactant mixture with turbulent temperature fluctuations. The diagram provides guidance on expected ignition behavior based on the thermo-chemical properties of the mixture and the flow/scalar field conditions. The analysis is an extension of the previous studies by Zeldovich (1980) and Sankaran et al. (2005) to combine the turbulent flow and scalar characteristics in terms of the characteristic Damköhler and Reynolds numbers of the system. The results of this work provided a more unified and comprehensive understanding of the physical and chemical mechanisms controlling ignition characteristics compared to the existing experimental maps in previous studies (Mansfield and Wooldridge, 2014; Meyer and Oppenheim, 1971), which were solely based on the ignition delay sensitivity.

It was recognized that the Zeldovich-Sankaran criterion includes the ignition delay sensitivity, $d\tau_{ig}/dT$, as a critical factor. Therefore, the traditional regime characterization based on the Damköhler and Reynolds numbers (such as those for turbulent premixed combustion regimes), which were based on time scales only, was not sufficient to describe the transitions between weak and strong ignition phenomena, and the introduction to the sensitivity parameter, *K*, was necessary. The regime diagram further showed how turbulence characteristics would affect the Zeldovich-Sankaran criterion based on the Kolmogorov theory of homogeneous isotropic turbulence.

The Zeldovich-Sankaran criterion indicates there is a region where mixtures with high-*K* values or high thermal sensitivity are more susceptible to weak ignition. Such conditions are reached with hydrogen/oxygen mixtures at low temperatures and high pressures. Therefore, the theory serves as a reasonable argument that the ignition delay discrepancies observed in syngas mixtures at low temperatures may be attributed to the ignition front propagation triggered by the local temperature peaks.

As for the turbulent combustion modeling implications, the proposed ignition regime diagram serves as a general guideline to identify whether the combustion processes are ignition-controlled versus flame-propagation-controlled. Many turbulent premixed combustion closure models inherently assume flame-dominant combustion mode, and it is hoped that the present study provides a metric to assess the validity of these models, as a supplement to the commonly-used Borghi diagram (Peters, 2000).

As a final remark, for higher hydrocarbon fuels that are known to exhibit the negative temperature coefficient (NTC) behavior, there is a broad range of intermediate temperature conditions at which the *K* value is expected to be low, thus promoting strong ignition. Weak ignition and associated front propagation behavior at NTC conditions have recently been studied (Yoo et al., 2011; Gupta et al., 2013; Kim et al., 2014; Mansfield et al., 2015). Further work is needed in order to validate the proposed regime diagram and ignition criterion for such complex fuels. Moreover, the sensitivity of the ignition characteristics to different uniform mixture compositions as well as the level of composition fluctuations would introduce further complexities to the problem. Additional detailed computational investigations using direct numerical simulations are underway.

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REFERENCES

Dryer, F.L., Wooldridge, M.S., Peterson, E. L., McDonell V.G., Im, H.G. October 21, 2014. Panel discussion: Ignition delay issue. United States Department of Energy University Turbine Systems Research (UTSR) meeting.

Gupta, S., Im, H.G., and Valorani, M. 2013. Analysis of n-heptane auto-ignition characteristics using computational singular perturbations. *Proc. Combust. Inst.*, 34, pp. 1125–1133.

Ihme, M. 2012. On the role of turbulence and compositional fluctuations in rapid compression machines: Autoignition of syngas mixtures. *Combust. Flame*, 159, pp. 1592-1604.

Javed, T., Es-sebbar, E., Jaasim, M., Badra, J., Im, H.G., Farooq, A. 2015. Interpreting low-temperature shock tube ignition delay data. *15th European Combustion Meeting*, Budapest, Hungary, March 30 – April 2.

Kee, R., Rupley, F., and Miller, J. 1989. CHEMKIN-II: a fortran chemical kinetics package for the analysis of gas-phase chemical kinetics. *SAND89-8009*, Sandia National Laboratories.

Kim, S.O., Luong, M.B., Chen, J.H., Yoo, C.S. 2014. A DNS study of the ignition of lean PRF/air mixtures with temperature inhomogeneities under high pressure and intermediate temperature. *Combust. Flame (in press)*, DOI: 10.1016/j.combustflame.2014.09.001.

Lavoie, G.A., Martz, J.B., Wooldridge, M.S., Assanis, D.N. 2010. A multi-mode combustion diagram for spark assisted compression ignition. *Combust. Sci. Tech.*, 157 (6):1106-1110.

Li, J., Zhao, Z., Kazakov, A., Chaos, M., Dryer, F.L., Scire, J.J. 2007. A comprehensive kinetic mechanism for CO, CH_2O and CH_3OH combustion. Int. J. Chem. Kinet., 39, pp. 109–136.

Lieuwen, T., McDonell, V., Santavicca, D., SattelMayer, T. 2008. Burner Development and Operability Issues Associated with Steady Flowing Syngas Fired Combustors. *Combust. Sci. Technol.*, 180(6), pp.1169–1192.

Lieuwen, T.C., Yang, V. (Eds), 2013. *Gas Turbine Emissions*, Cambridge Aerospace Series, Book 38, Cambridge University Press, Ch. 1-2.

Linan, A., and Williams, F.A. 1993. Fundamental aspects of combustion. *Oxford University Press*.

Mansfield, A.B., and Wooldridge, M.S. 2014. High-pressure low-temperature ignition behavior of syngas mixtures. *Combust. Flame,* 161 (9), pp. 2242-2251.

Mansfield, A.B., Wooldridge, M.S., Di, H., He, X. 2015. Low-temeperature ignition behavior of iso-octane. *Fuel*, 139, 79-86.

Meyer, J.W., and Oppenheim, A.K. 1971. On the shock-induced ignition of explosive gases. *Proc. Combust. Inst.*, 13, pp. 1153–1164.

Mittal, G., Sung, C.J., and Yetter, R.A. Autoignition of H_2/CO at elevated pressures in a rapid compression machine. *Int. J. Chem. Kinet.*, 38, pp. 516-529.

Oran, E.S., Young, T.R., Boris, J.P., Cohen, A. 1982. Weak and strong ignition. I. Numerical simulations of shock tube experiments. *Combust. Flame*, 48, pp. 135-148.

Oran, E.S., and Borris, J.P. 1982. Weak and Strong Ignition. II. Sensitivity of the Hydrogen-Oxygen System. *Combust. Flame*, 48, pp. 149-161.

Peters, N. 2000. *Turbulent Combustion*, Cambridge University Press, Ch. 2.

Petersen, E.L., Kalitan, D.M., Barrett, A.B., Reehal, S.C., Mertens, J.D., Beerer, D.J., Hack, R.L., McDonell, V.G. 2007. New syngas/air ignition data at low temperature and eevated pressure and comparison to current kinetic models. *Combust. Flame*, 149, pp. 244–247.

Richards, G.A., McMillian M.M., Gemmen, R.S., Rogers, W.A., Cully, S.R. 2001. Issues for low-emission, fuel-flexible power systems. *Prog. Energy Combust.*, 27(2), pp.141–169.

Sankaran, R., Im, H.G., Hawkes, E.R., Chen, J.H. 2005. The effects of non-uniform temperature distribution on the ignition of a lean hydrogen-air mixture. *Proc. Combust. Inst.*, 30, pp. 875–882.

Tennekes, H., and Lumley, J.L. 1972. A first course in turbulence. *MIT press*, Cambridge, MA.

United States Department of Energy. 2009. Hydrogen from Coal Program - Research, Development and Demonstration Plan. http://fossil.energy.gov/programs/fuels/publications/programplans/2009_Draft_H2fromCoal_Sept30_web.pdf

Uygun, Y., Ishihara, S., and Olivier, H. 2014. A high pressure ignition delay time study of 2-methylfuran and tetrahydrofuran in shock tubes. *Combust. Flame*, 161, pp. 2519-2530.

Walton, S.M., He, X., Zigler, B.T., Wooldridge, M.S. 2007. An experimental investigation of the ignition properties of hydrogen and carbon monoxide mixtures for syngas turbine applications. *Proc. Combust. Inst.*, 31, 3147–3154.

Wu, H., and Ihme, M. 2014. Effects of flow-field and mixture inhomogeneities on the ignition dynamics in continuous flow reactors. *Combust. Flame*, 161, pp. 2317-2326.

Yoo, C.S., Lu, T., Chen, J.H., Law, C.K. 2011. Direct numerical simulations of ignition of lean n-heptane/air mixture with temperature inhomogeneities at constant volume: Parametric study. *Combust. Flame*, 158, pp. 1727-1741.

Zeldovich, Y.B. 1980. Regime classification of an exothermic reaction with nonuniform initial conditions. *Combust. Flame*, 39, pp. 211–214**.**

Table 1: Details of the experimental cases of Mansfield and Wooldridge (2014) and corresponding regime diagram parameters

Mixture composition[a] (% vol)					**Assigned thermo. state**		**Ignition behavior[b]**	**0D ignition delay, τ_{ig} (msec)**	**Ignition delay sensitivity, $\lvert d\tau_{ig}/dT \rvert$ (ms/K)**	**Flame time scale, τ_f (μs)**	**K^c**	**Da_l^c**	**K^2/Da_l**	**Re_l**	**Da_λ**
H_2	**CO**	**O_2**	**N_2**	**CO_2**	**P (atm)**	**T (K)**									
5.1	7.3	12.5	63.0	12.0	15.5	983	M	23.0	0.57	21.40	4.04	8.69	1.88	263.16	1.35
5.1	7.3	12.7	73.8	1.1	9.2	1020	M	9.4	0.25	4.21	6.3	21.27	1.87	140.84	4.09
5.1	7.3	12.6	73.8	1.2	9.7	1033	M	6.7	0.18	3.76	5.58	29.85	1.04	144.92	5.68

5.1	7.3	12.7	73.8	1.1	5.5	1043	M	3.7	0.14	1.50	9.13	54.05	1.54	80.96	12.50
5.1	7.3	12.6	63.1	11.9	4.4	962	W	55.0	1.63	10.15	10.9	3.64	32.5	78.72	0.85
1.7	2.4	20.8	70.4	4.7	4.4	1043	S	4.5	0.25	92.0	1.94	44.44	0.085	68.96	10.83
1.7	2.5	20.9	73.3	1.7	4.7	1012	S	12.0	0.63	425.0	1.39	16.67	0.12	75.18	3.95
1.7	2.4	20.8	69.9	5.2	3.0	995	S	26.0	1.03	166.0	2.48	7.69	0.8	50.37	2.08
1.7	2.4	20.8	74.8	0.3	2.4	992	S	10.0	0.86	87.30	4.59	20.0	1.05	39.21	5.89
1.7	2.6	20.8	74.8	0	3.1	1029	S	2.1	0.17	30.0	3.39	95.23	0.12	74.07	22.68

[a] Balance Ar

[b] Experimentally observed. M: mixed, W: weak, S: strong ignition.

[c] Assumed quantities: T’ ~ 10 K, u’ ~ 0.1 m/s, l ~ 2 cm

Figure 1: A schematic of combustion chamber with various physical length scales.

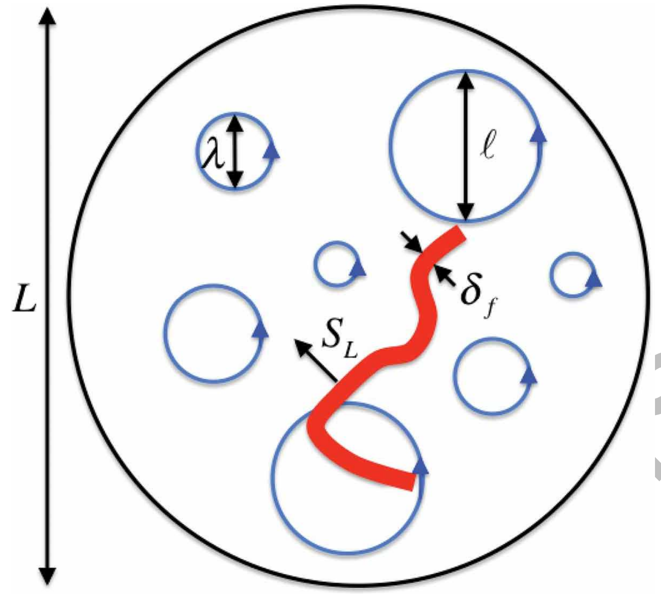


Figure 2: Regime diagram for strong and weak ignition for nearly homogeneous reactant mixture with temperature fluctuations.

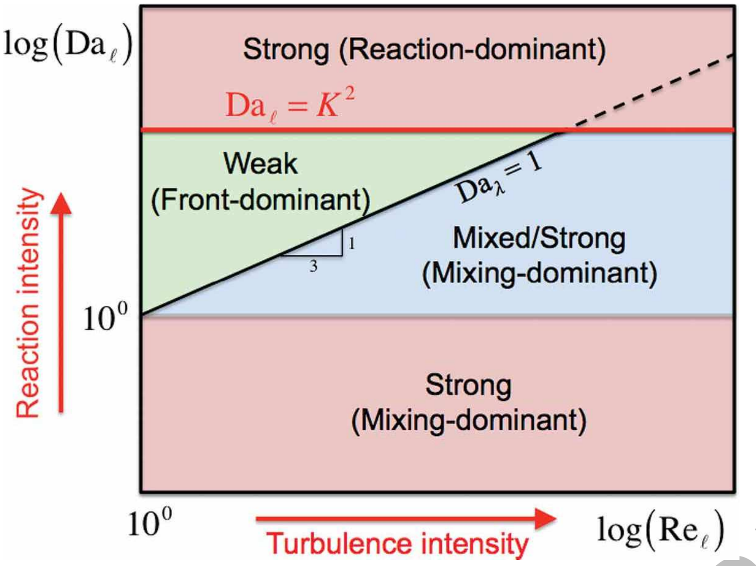
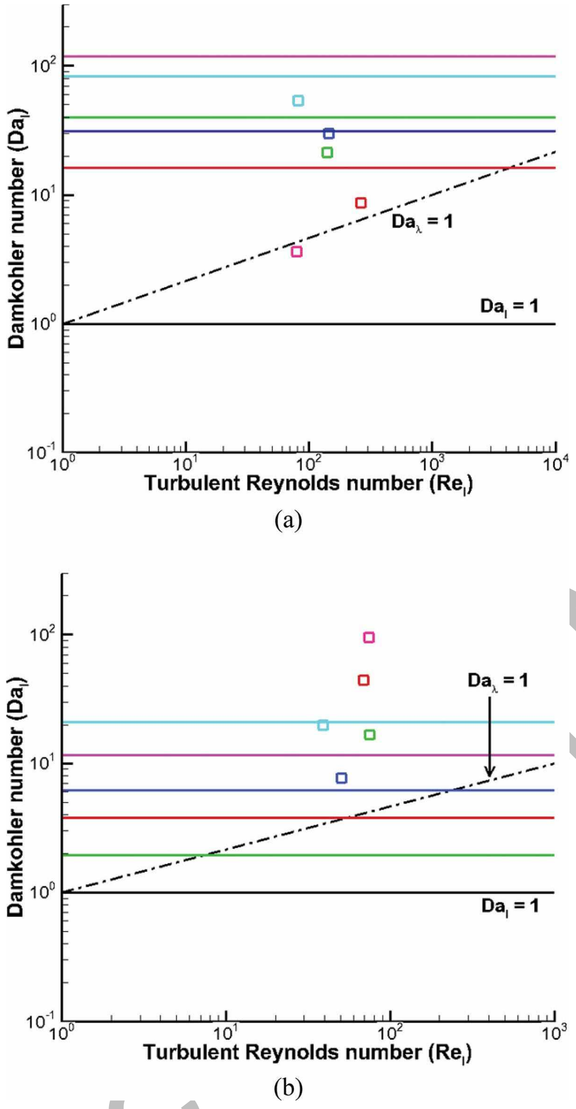


Figure 3: Location of (a) weak/mixed ignition cases 1-5 and (b) strong ignition cases 6-10 in red, green, dark blue, sky blue, pink, respectively, on the turbulent regime diagram.



(a)

(b)